5.5 Annual Mass Balance for Other Constituents

The U-236 values shown in Table XV were values actually measured on composited samples of dissolver product during the late 1980s. These values are the maximum values reported for uranium-236 and were determined by mass spectrometry.

Uranium-236 was included because it results in significant radiation exposures in aged material due to the presence of decay product, uranium-232 and its daughters, particularly thallium-208 which is short-lived with a high-energy (2.6 Mev) gamma emission.

5.6 Potential for Worker Exposure from Recycled Uranium

As the calculations in Section 2.4 indicated most of the effective dose equivalent exposures would be due to the uranium radionuclides (see Table III). Uranium-234, Because of its short half-life (2.45×10^{-5} years) compared to the half-lives (10^{-7} to 10^{-9} years) of the other uranium isotopes in ICPP product, uranium-234 is often the dose limiting radionuclide. Uranium-234 is significantly concentrated by the gaseous diffusion plants and then increased slightly more in a reactor through n, 2n reactions with uranium-235. Throughout the history of ICPP, the risk of exposure to radionuclides in final product was based on the uranium isotopes rather than the actinide or fission product radionuclide. As can be seen in Table III, the plutonium isotopes are at least an order of magnitude lower risk than the highest risk uranium isotope. High-enriched, high-burnup fuels have high concentrations of uranium-234, -235, and -236 which are the limiting isotopes in handling ICPP product.

The bioassay programs would pick up internal exposures to uranium. The uranium that was frequently observed was usually natural uranium from the environment and was not considered to be a problem at that level. The presence of uranium-234 or uranium-236 or of higher enrichments of uranium-235 would result in follow up to determine the extent of the dose and the source. In general, because of the monitoring for uranium isotopes, the risk of exposure to other constituents in ICPP product, was small.

5.7 Potential for Environmental Contamination from Recycled Uranium.

There was no risk of environmental contamination from ICPP recycled uranium product.

6.0 RESULTS AND CONCLUSIONS

6.1 Explanation of Mass Flow Paths and Contaminant Levels

Material shipped from the Idaho Chemical Processing Plant was sent to Y-12 and to Portsmouth for future processing. Smaller quantities were sent to Rocky Flats, Hanford and Los Alamos for criticality studies. This material was subsequently either returned to ICPP for cleanup or sent directly to Y-12 for processing prior to being shipped to Savannah River. Some is still believed to be in inventory at the

receiving site. Alpha contamination of material sent to Y-12 was below their specification. Beta contamination was four to five times their specification in shipments sent between 1953 and 1977. After 1977, the beta contamination was consistently below their specification.

6.2 Identification of Processes or Areas of Concern for Worker Exposure

Exposure to the product material was to the operations personnel who packaged the product and took samples, maintenance personnel who maintained the final product equipment, health physics personnel monitoring radiation exposures, and to the analytical personnel who analyzed the product samples. Prior to 1971, the product and the samples were liquids in the form of a concentrated uranyl nitrate solution in nitric acid. After 1971, the product and the samples were essentially pure uranium trioxide powder and particles. The highest risk was due to the uranium isotopes compared to the other actinides or to technetium.

6.3 Identification of Processes or Areas of Concern for Environmental Impact

Environmental impact statements have been prepared for all phases of the processes at ICPP. No areas of concern with respect to any of the processes for the handling of final product were identified.

6.4 Discussion of Data Sources and Confidence Levels

Three different sources of shipment data were used to determine the amount of product that was shipped from the ICPP. The data was taken from DOE/OR-859 (The Egli Report), a collection of monthly ICPP production reports, and a compilation of shipments by date and RIS codes made by an accountability manager. The combination of this data appears to provide an accurate assessment of the shipments, particularly in the absence of a large fraction of the 741 forms from one of the early site contractors. A subsequent check at Y-12 indicated that the shipping records that they had, matched the tables of shipments made through the years as documented by the accountability personnel at ICPP.

Original analytical data was sent to a records repository in Seattle and then subsequently destroyed. Compiled data from some of the more recent shipments is available for transuranics in the dissolver product. Additional data is given in the Egli Report (DOE/OR-859) based on information developed at Y-12 when analyses were completed on uranium product sent to Y-12.

Analyses for technetium do not exist at ICPP. Technetium was never a concern in the product and as such was never requested. Because it was not a concern, an analytical method was not developed for the separation and analysis of technetium until 1998.

Because original records do not exist for much of the data, confidence in the data is not as high as it would be with a complete, original data set. The use of original, complete data sets would produce the highest level of confidence. But, because a

significant amount of data has been lost or destroyed, this level of confidence is not possible. Ideally, the backup information normally associated with the shipping documents would include the analytical chemistry data, description of the material in the shipment, shipment packaging, etc. This means that other sources of data must be identified and utilized. The confirmation that the records that Y-12 have matches the tabular shipping data gives confidence that these are equivalent to original data.

What is available are several different data sets that were produced for different reasons for different groups. The fact that this data is quite consistent provides confidence that even though the original data is lost, the data that has been preserved as a secondary source of data is consistent and therefore increases confidence in these secondary sources. A paragraph in the Egli report indicates that transuranic alpha contamination was always below the receiver's specification. In the early years, the beta contamination was four to five times the specification but from 1977 on, the beta activity was below the specification. Utilizing this information allows one to back calculate the alpha emitting materials present in the product. This allows one to estimate, with confidence, the amount of transuranics in the ICPP product.

Estimates of the range of the constituents content in the three fuel types was made by using the data that was calculated based on the alpha specification and on the values calculated from the DFS and the ORIGEN2 results. As indicated earlier the "most probable" constituent levels are based on the data presented in the Egli report. The Egli data is based on analytical results of product received at Y-12. The data from the ORIGEN2 calculations combined with the experimental DFs both have large uncertainties which are probably over estimating the contaminant concentrations.

The ranges are shown in Table XVI. For plutonium the range is very large for aluminum and zirconium fuels. For stainless steel, the range is actually quite narrow, probably due to the fact that fuel had a lower burnup, and because the plutonium isotopic distribution is essentially only the plutonium-239 isotope.

The range for neptunium is also close together again probably because there is only a single isotope produced.

The technetium-99 data is only based on the ORIGEN2 calculations and the total beta DF. Because it is known that the isotope that affected the beta ratio data was primarily ruthenium-106 rather than technetium-99, the entire range probably significantly over estimates the technetium-99 concentration.

Table XVI
Ranges of Contaminants

	Aluminum	Zirconium	Stainless Steel
Pu	0.022 ppb - 3 ppm	0.001 ppb - 300 ppb	21 ppb - 35 ppb
Np-237	1.2 ppm - 2.5 ppm	1.6 ppm - 4 ppm	7.4 ppb - 31 ppb
Tc-99	1.0 ppb - 1.1 ppb	1.7 ppb - 1.8 ppb	0.018 ppb - 002 ppb

6.5 Conclusions

The Idaho Chemical Processing Plant produced 32.053 MTU product as the result of processing spent nuclear fuel. Of that amount 25.773 MTU was shipped to Y-12 and 4.076 MTU was sent to Portsmouth. In addition, 0.219 MTU was sent to Rocky Flats, 0.047 MTU was sent to PNNL and 0.168 MTU was sent to Los Alamos. All of the small quantities (less than one metric tonne) were used in criticality experiments. In addition to the material that was shipped off site, there is still in inventory 1.770 MTU of uranium product at ICPP.

There was a total of 30.283 MTU shipped which contained 0.112 grams of plutonium, 24.70 grams of neptunium and 0.028 grams of technetium -99. Y-12 received 0.025 grams of plutonium, 24.34 grams of neptunium and 0.028 grams of technetium. Portsmouth received 0.087 grams of plutonium, 0.127 grams of neptunium and 0.0001 gram of technetium-99. These numbers are our best estimates for this data. They are based on alpha ratio data from analytical measurements at Y-12 and ORIGEN2 code calculations which provided the radionuclide distribution from that data, a calculation can be made that provides an estimate of the transuranic radionuclides present in ICPP product.

Radiologically the dose potential associated with ICPP product and the equipment associated with producing, packaging, and analysis of the product was primarily due to the uranium isotopes in the product and not due to the higher actinides or the technetium-99. The uranium isotopes that limited the potential dose were uranium-234 or uranium-235. In some cases, high levels of uranium-236 could become a problem after the ingrowth of uranium-236 daughters - particularly thallium-208. The dose to workers from plutonium isotopes and neptunium-237 while handling ICPP product was at least two orders of magnitude less than that from the uranium isotopes.

In general, because the dose potential from ICPP product was limited by uranium isotopes, operations were conducted in a manner to confine the product and minimize the risk to workers. Radiation monitoring focused on the alpha contamination for worker protection. In addition, added protection was provided through working with the material in glove boxes and hoods. While there were low level exposures and internal exposures through the years, they did not result in any doses in excess of the allowable limits.